



IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re the application of
Suguru Ito et al
Serial No. 10/089,104
Filed: April 15, 2002

Group Art Unit: 1713
Primary Examiner:
Robert D. Harlan

For: RUBBER, RUBBER COMPOSITION, AND CROSSLINKED OBJECT

DECLARATION UNDER 37 C.F.R. 1.132

Honorable Commissioner of Patents and Trademarks
Washington, D.C. 20231

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Sir:

I, Shigeru Fujita, of c/o the Research and Development Center, Zeon Corporation, 2-1, Yako 1-chome, Kawasaki-ku, Kawasaki-shi, Kanagawa, 210-9507 Japan, being duly sworn, declare and state:

THAT I am by profession a research chemist having had a major in organic synthetic chemistry and been awarded a master's degree from the post-graduate course of the Faculty of Industrial Chemistry, Kantogakuin University in March 1993.

THAT I have been employed since April 1993 by Zeon Corporation of 6-1, Marunouchi 2-chome, Chiyoda-ku, Tokyo, 100-0005 Japan and engaged in research on properties of polymers, especially hydride rubber and hydrogenated nitrile-butadiene rubber in the Research & Development Center, Elastomer Laboratory of the same company.

THAT I took over the researches on development of hydrogenated nitrile-butadiene rubber and prosecution of the above-identified U.S. patent application from the applicants of the above-identified U.S. patent application, and were informed

of the invention disclosed in said U.S. patent application (which invention is hereinafter referred to as "present invention") from the applicants, and hence, I am fully familiar with the present invention.

THAT, in order to show that the rubber of the present invention gives a crosslinked rubber product exhibiting greatly reduced swelling in oil, improved resistance to rancid gasoline, and greatly reduced hardening in oil, which are believed to be unpredictable from Oyama et al U.S. Patent No. 4,643,938, the following comparative experiments have been carried out under my supervision.

Comparative Experiments

I. Experiment I (Test of copolymer rubbers disclosed in Oyama et al)

Oyama et al disclose acrylonitrile/butadiene copolymer rubbers, and, more specifically, in the working examples of Oyama et al, an acrylonitrile/butadiene copolymer rubber (E), acrylonitrile/butadiene/hydrogenated butadiene copolymer rubbers (A, B, C and D) and acrylonitrile/butadiene/methyl acrylate copolymer rubbers (F and G).

I prepared acrylonitrile/butadiene copolymer rubbers having the same compositions as those of copolymer rubbers A, B, C, D and G disclosed in Oyama et al, and crosslinked these acrylonitrile/butadiene copolymer rubbers to produce crosslinked rubber products by the same procedures as described on page 16, lines 9-27 (Examples 1-7, Comparative Examples 1-7) of the specification of the above-identified U.S. patent application. Oil resistances [i.e., (i) volume change at immersion in fuel D, (ii) hardening at immersion in fuel D containing a condensed aromatic mixture, and (iii) resistance to rancid gasoline] of the crosslinked rubber products were measured by the methods

described in the paragraphs on pages 15 and 16 of the specification of the above-identified U.S. patent application. The results are shown in Table I, below.

Note, copolymer rubbers E and F disclosed in the working examples of Oyama et al had compositions quite different from that of the copolymer rubber of the present invention, and therefore, tests of oil resistances of these copolymer rubbers E and F were not conducted in these Comparative Experiments.

II. Experiment II (Test of additional comparative copolymer rubbers)

Comparative copolymer rubbers (Comparative Examples 7 and 8) having a composition (shown in Table I, below), which are similar to that of the copolymer rubber of the present invention in that the comparative copolymer rubbers contain 1,3-butadiene units (a), saturated 1,3-butadiene units (b), acrylonitrile units (c) and other monomer (butyl acrylate) units (d), but are different from the copolymer rubber of the present invention in that the content (35% by weight) of acrylonitrile units (c) in the comparative copolymer rubbers is smaller than that in the copolymer rubber of the present invention.

The comparative copolymer rubbers were crosslinked, and oil resistances thereof were measured by the same methods as in Experiment I. The results are shown in Table I, below.

For comparison, the results in Examples 1-8 and Comparative Examples 1-6 shown in Tables 1 and 2 on pages 18 and 19 of the specification of the above-identified U.S. patent application are shown in Table I, below.

Table I

Example No.	Examples								Comparative Examples						Examples of Oryama '938				Comp. Ex.*5			
	1	2	3	4	5	6	7	8	1	2	3	4	5*6	6	A	B	C	D	G	7	8	
Monomer unit *1 (wt.%)																						
(a)BD	7	7	5	5	6	6	6	6	6	6	7	5	6	8	18.9	6.3	5.5	34.6	30	25	5	
(b)HBD	28	19	29	32	38	34	34	34	50	47	41	50	9	39	44.1	56.7	49.5	28.4	-	20	30	
(c)AN	44	44	45	42	44	42	42	42	44	44	44	25	50	38	37	37	45	37	35	35	35	
(d)BA	21	30	21	21	-	-	-	-	-	3	8	20	35	15	-	-	-	-	35	20	30	
Ip	-	-	-	-	12	18	-	-	-	-	-	-	-	-	-	-	-	-	-	-	-	
St	-	-	-	-	-	-	18	18	-	-	-	-	-	-	-	-	-	-	-	-	-	
(d) (mol%)	10	15	10	10	10	15	10	10	0	1	4	9	18	7	0	0	0	0	18	10	15	
(a)H(b)(wt.%)	35	26	34	37	44	40	40	40	56	53	48	55	15	47	63	63	55	63	30	45	35	
Vol change *2	32	31	35	38	38	42	42	43	35	35	36	70	25	48	41	42	33	42	57	51	54	
Difference *3																						
In hardness	0	0	2	2	2	0	0	0	20	17	7	0	0	8	6	19	20	4	0	0	0	
Cracks *4	N	N	N	N	N	N	N	N	N	N	N	N	N	N	P	N	N	P	P	P	N	

Notes, *1 Monomer unit composition of rubber (wt. %): (a) BD: 1,3-Butadiene units, (b) HBD: Hydrogenated butadiene units, (c) AN: Acrylonitrile units,

(d) BA: Butyl acrylate units, Ip: Isoprene units, St: Styrene units

*2 Volume change in fuel D immersion test

*3 Difference in hardness in Hardening test

*4 Presence of cracks in random gasoline resistance test, N: Cracks were not present, P: Cracks were present

*5 Comparative Examples 7 and 8: Newly added Comparative Examples

*6 Crosslinked product in Comp. Ex. 5 exhibits good oil resistances, but has very poor tensile strength (10 MPa).

III. Evaluations of Experimental Results

- (1) Butadiene/acrylonitrile/butyl acrylate copolymer rubber G in Oyama et al

Copolymer rubber G disclosed in Oyama et al is different from the copolymer rubber of the present invention in that copolymer rubber G does not contain saturated butadiene units (b), and copolymer rubber G contains a larger amount (30% by weight) of butadiene units (a), and a smaller amount (35% by weight) of acrylonitrile units (c), than the copolymer rubber of the present invention.

Copolymer rubber G of Oyama et al gives a crosslinked product which exhibits an undesirably large volume change (57%) at immersion in fuel D and in which cracks occur on the surface at rancid gasoline test.

- (2) Butadiene/saturated butadiene/acrylonitrile copolymer rubbers A, B, C and D in Oyama et al

Copolymer rubbers A, B, C and D in Oyama et al are different from the copolymer rubber of the present invention in that at least one of the amounts of butadiene units (a), saturated butadiene units (b) and acrylonitrile units (c), is different from the copolymer rubber of the present invention, and the sum of (a)+(b) in each of copolymer rubbers A, B, C and D of Oyama et al is larger than that in the copolymer rubber of the present invention. Further, copolymer rubbers A, B, C and D of Oyama et al do not contain monomer units (d), other than monomer units (a), (b) and (c).

Copolymer rubbers A, B, C and D of Oyama et al give crosslinked products exhibiting undesirably large hardening at immersion in fuel D containing a condensed aromatic mixture, or having poor resistance to rancid gasoline.

- (3) Butadiene/saturated butadiene/acrylonitrile/butyl acrylate copolymer rubbers (Comp. Ex. 7 and 8)

The copolymer rubbers in Comparative Examples 7 and 8 have a composition, which is similar to that of the copolymer rubber of the present invention in that the comparative copolymer rubbers contain 1,3-butadiene units (a), saturated 1,3-butadiene units (b), acrylonitrile units (c) and other monomer (butyl acrylate) units (d), but is different from the copolymer rubber of the present invention in that the comparative copolymer rubbers contain smaller amounts (35% by weight) of acrylonitrile units (c).

The copolymer rubber in Comparative Example 7 gives a crosslinked product exhibiting a large volume change at immersion in fuel D and having poor resistance to rancid gasoline. The copolymer rubber in Comparative Example 8 gives a crosslinked product exhibiting a large volume change at immersion in fuel D.

- (4) Butadiene/saturated butadiene/acrylonitrile/butyl acrylate (or isoprene or styrene) copolymer rubbers of the present invention (Examples 1-8)

The copolymer rubbers of the present invention (Examples 1-8) give crosslinked products exhibiting (i) a greatly reduced volume change at immersion in fuel D, (ii) a greatly reduced hardening at immersion in fuel D containing a condensed aromatic mixture, and (iii) an improved resistance to rancid gasoline.

I, the undersigned declarant, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001, of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

This twentieth day of January, 2004

Shigeru Fujita

Shigeru Fujita